**Article title:** Optical dephasing of paramagnetic ions: Er3+ : YLiF4 — experiments and computer simulations

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**Context**

Spectral diffusion – the change in frequency of a transition with time - is seen in studies of the optical transitions of rare earth ions in crystals. It is often caused by fluctuating magnetic fields due to electron or nuclear spin flips in the environment, driven by a variety of processes (e.g. resonant spin flip-flops, interaction with phonons).

In the frequency domain, spectral diffusion causes the broadening of the transition, meaning it is hard to measure the “true” homogeneous linewidth (i.e. without time-dependent broadening). Spectral diffusion can also be seen in the time domain, in photon echo experiments, as a non-exponential decay of the ensemble coherence over time. Other workers (e.g. Mims, ref [4], DeVoe ref[6]) have investigated this decay. Mims proposed a (now very well known) semi-empirical mathematical model for the decay shape, which people have used to explain several materials.

**Purpose**

To understand the spectral diffusion processes causing a non-exponential photon echo decay in 0.02%Er doped YLiF4, in high field. The idea is to complement other studies, done on rare earths with much lower electronic magnetic moment (e.g. Pr). Here, most common spectral diffusion mechanisms seen in other crystals are absent, leaving, they say, only nuclear spin flip-flop processes of the F spins.

**Approach**

The authors compare experimental results with computer simulations. The experiments were optical photon echoes on the 4I15/2-4F9/2 transition, performed at very low temperatures (1.5 K) and in high magnetic fields (3-5 T) to eliminate spectral diffusion due to phonons and electron spin flips.

In their computer simulation, they use the Monte Carlo approach of DeVoe(ref [6]). They model the ~1000 flourine (F) ions in the YLiF4 lattice directly surrounding an Er ion. The F ions interact with the Er, and each other, via magnetic dipole-dipole interactions. Rates for mutual F ion spin flips are calculated using Fermi’s golden rule. In each iteration, they calcuate the fluctuation in the magnetic field at the Er site, repeated for many (103-105) Er sites. This gives them the coherence in the ensemble.

**Contribution**

Compared to, say, Pr, Er crystals have a large “frozen core” of nuclear spins whose frequency is detuned from the bulk F spins by the large Er magnetic moment, so they flip slowly.

The authors showed that this means it is actually the more distant spins that are the major source of spectral diffusion (whereas other systems, (Pr) are dominated by the frozen core). Their computer model reproduced the shape of the experimental spectra, although it predicted a stronger correlation in spin flip rate with distance than the data suggested. They estimate a nuclear spin correlation time of 200 us, in agreement with measurements in LaF3. Their results can be applied to other Er crystals in high field and low temperature.

**Relevance**

Spectral diffusion is important to understand for my work on quantum memories, where it decreases the storage time. I’m familiar with some work (refs 4-6) on modelling spectral diffusion, but this is the first paper I’ve seen that looks at Er, which we want to start using for memories. They show that the dominant dephasing mechanisms are different from in Pr/Eu crystals. It would be interesting to apply this model to our Er:Y2SiO5 high field results, and see if it fits.

**Quality**

Writing is a little wordy, but well structured. The paper is pretty clear and rigorous. Their justification that electron spin flips are not significant (i.e. no field dependence at high fields) seems reasonable, although it surprises me since the concentration is so high (0.02%). Their computer simulations are simple and do make some assumptions, but they check they are reasonable, e.g. the lattice size.

**Questions/Directions**

* They are working with the 4F9/2 excited level – how do its properties compare to the more commonly used 4I13/12? I’m assuming the lifetime is short, coherence time also?
* Coherence time is only 10 us, which is super short. Why? The high magnetic moment of the F spins?
* I didn’t realise the exponent in the Mims equation could go above 2. Need to find out what conditions cause this – is it the correlations in rate with distance?
* We’ve never had much luck with getting good predictions from Fermi’s golden rule – what is the difference here?
* I should try out this model on data that we have.
* What similar work has been published since? Look at [T. Böttger et al., Phys. Rev. B **79**, 115104 (2009)] – also about long coherence times in Er.